RADIATION PHYSICS NOTE 99

Note on Leaking Radium Sources

November 1992

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In the process of investigating the source of a minor contamination in the large source safe, a number of ^{226}Ra radioactive sources[1] were found to be leaking. To characterize the nature of the contamination, each source was sealed in a quart aluminum can along with a standard cloth wipe, and stored in a ziplock bag over night. The cloth wipe traps and collects any ^{222}Rn daughters if Rn is leaking from the sources. The following morning the "hottest" wipe[2] was taken to 14 Shabbona and counted for both beta, gamma and alpha activity separately on the sample changer for 8 hours in one minute intervals. The results are shown in Fig. 1.

The decay chain for ^{226}Ra is shown in Fig. 2. Note in particular that in the alpha decay through ^{222}Rn and ^{218}Po, ^{214}Pb is made and subsequently beta decays with a $T_{1/2}$ of 26.8 minutes to ^{214}Bi. This then decays by beta, gamma emission (with a $T_{1/2}$ of 19.7 minutes) to ^{214}Po, which goes to ^{210}Pb by alpha decay with a very short half-life.

During the times appropriate to the above counting period and intervals, the general equations of radioactive growth and decay[3] for the ^{226}Ra series, can be shortened. The population of ^{214}Bi nuclei can be written as:

$$N_{Bi} = (N_{Pb})_0 \frac{\lambda_{Pb}}{\lambda_{Bi} - \lambda_{Pb}} \left(e^{-\lambda_{Pb} t} - e^{-\lambda_{Bi} t} \right), \qquad (1)$$

where $(N_{Pb})_0$ is the initial pure population of the parent ^{214}Pb nuclei, and the decay constants are $\lambda_{Pb} = 0.0259$ min^{-1} and $\lambda_{Bi} = 0.0352$ min^{-1}. At any time t, therefore, the counting rate data for beta, gamma emission as measured with the sample changer can be written:

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[1] These sources are 1" diameter by 5/16" thick disc source, with about 15 micrograms of Ra, obtained in 1970 from Nuclear Associates, Inc., Westbury, NY.

[2] The wipe that was sealed up with source 226-1.1-6.

[3] R. Evans, The Atomic Nucleus, Ch. 15, McGraw-Hill, NY, 1955

$$N_{data} = (N_{Pb})_0 \left[e^{-\lambda_{Pb} t} + C \frac{\lambda_{Pb}}{\lambda_{Bi} - \lambda_{Pb}} \left(e^{-\lambda_{Pb} t} - e^{-\lambda_{Bi} t} \right) \right], \quad (2)$$

where C represents the contribution to the observed counting rate from ^{214}Bi build-up and decay; its value essentially arises from the difference in efficiency for detection of ^{214}Pb and ^{214}Bi activity. This difference is due to the much reduced efficiency with which the sample changer (proportional counter) detects gamma rays relative to betas. It should be noted that the alpha counting data is governed by the same expression (2) because of the extremely short half-life in the decay of ^{214}Po to ^{210}Pb.

The data shown in Fig. 1 were fit to expression (2). The results are shown in Fig.3. For beta, gamma emission the constant C is 0.184, while for alpha emission it is 0.25. As observed the fit in both cases is excellent. For times greater than 300 minutes, the data (which are larger than the normal sample changer backgrounds) are consistent (see Fig. 4) with a lifetime on the order of days, although the statistical accuracy is poor. It should be noted that the ^{222}Rn parent has a half-life of 3.8 days.

A wipe sample, obtained in a manner similar to that discussed above, was analyzed by gamma-ray spectroscopy using the HpGe detector at the Activation Analysis Laboratory. Two conclusions can be drawn from this analysis. First, the only radionuclides observed were ^{214}Pb and ^{214}Bi. Apparently the ^{226}Ra source did not lose any solid material but simply fractured enough to allow radon gas to escape. And second, the ratio of ^{214}Bi to ^{214}Pb activity, based on a two hour counting period starting 17 minutes after a 24 hour "irradiation" (i.e., the time during which the wipe was sealed up in the can with the leaking source) was found in the analysis to be 1.79.

From (1), above, the ratio of activities can be written as:

$$\frac{\lambda_{Bi} N_{Bi}}{\lambda_{Pb} N_{Pb}} = \frac{\lambda_{Bi}}{\lambda_{Bi} - \lambda_{Pb}} \left(1 - e^{-(\lambda_{Bi} - \lambda_{Pb}) t} \right). \quad (3)$$

Based on this expression, the measured ratio of activities (1.79) would be expected at a time about 70 minutes after the end of "irradiation." This time is very close to the average of the total time after the wipe was removed from the aluminum can during which the analysis was performed, and thus the measured ratio of activities is in excellent agreement with what would be expected based on the equations of radioactive growth and decay.

We wish to thank the Rad Con team for their assistance with this investigation.

Figures

1. Counts per minute as a function of time after removing the wipe from the Al can. Each point represents a 1 minute counting time.

2. Decay chain of ^{226}Ra. The times shown are half-lives.

3. Counts per minute as a function of time for both beta, gamma emission (top) and alpha decay (bottom). The thick black line represents a fit based on eq. (2) in text, with C=0.184 for top curve and 0.24 for the bottom curve.

4. Counts per minute at counting times greater than 340 minutes, compared to expected time dependence for a decay with a half-live of the order of days.

Figure 1

RADIUM SOURCE 226-1.1-6 WIPE RESULTS

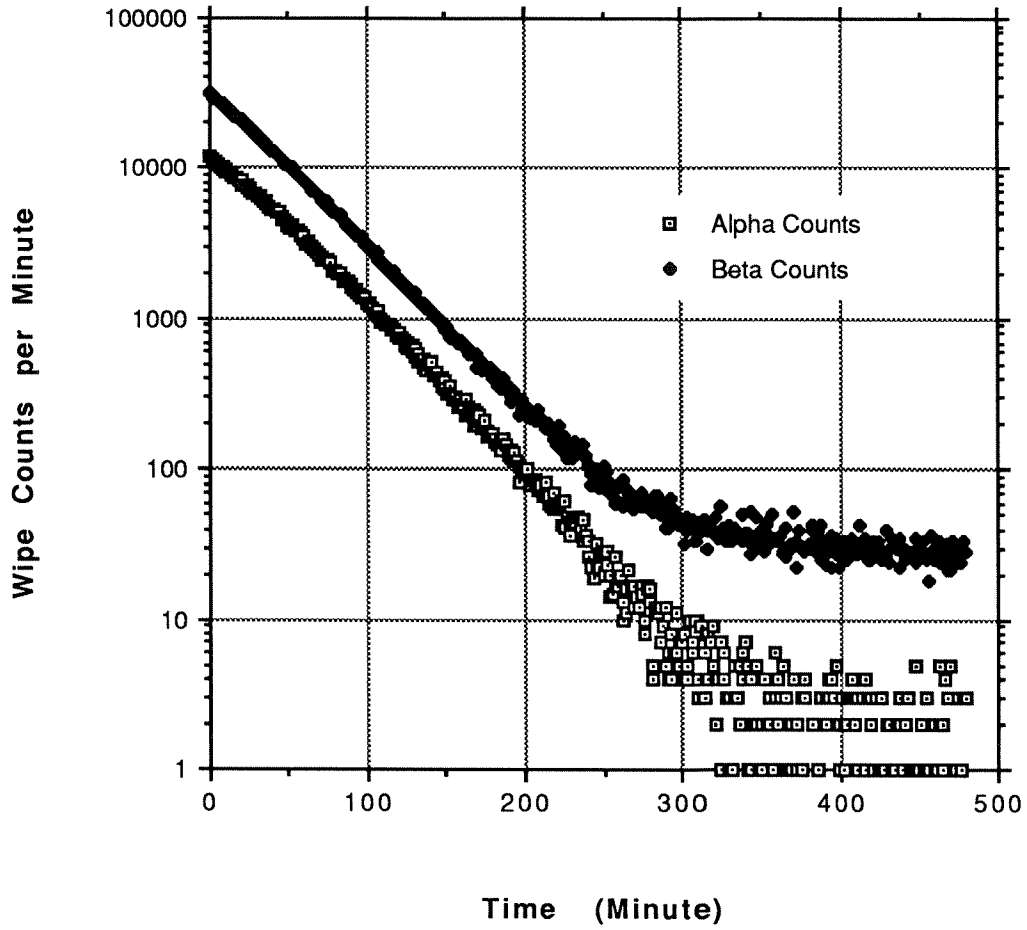


Figure 2

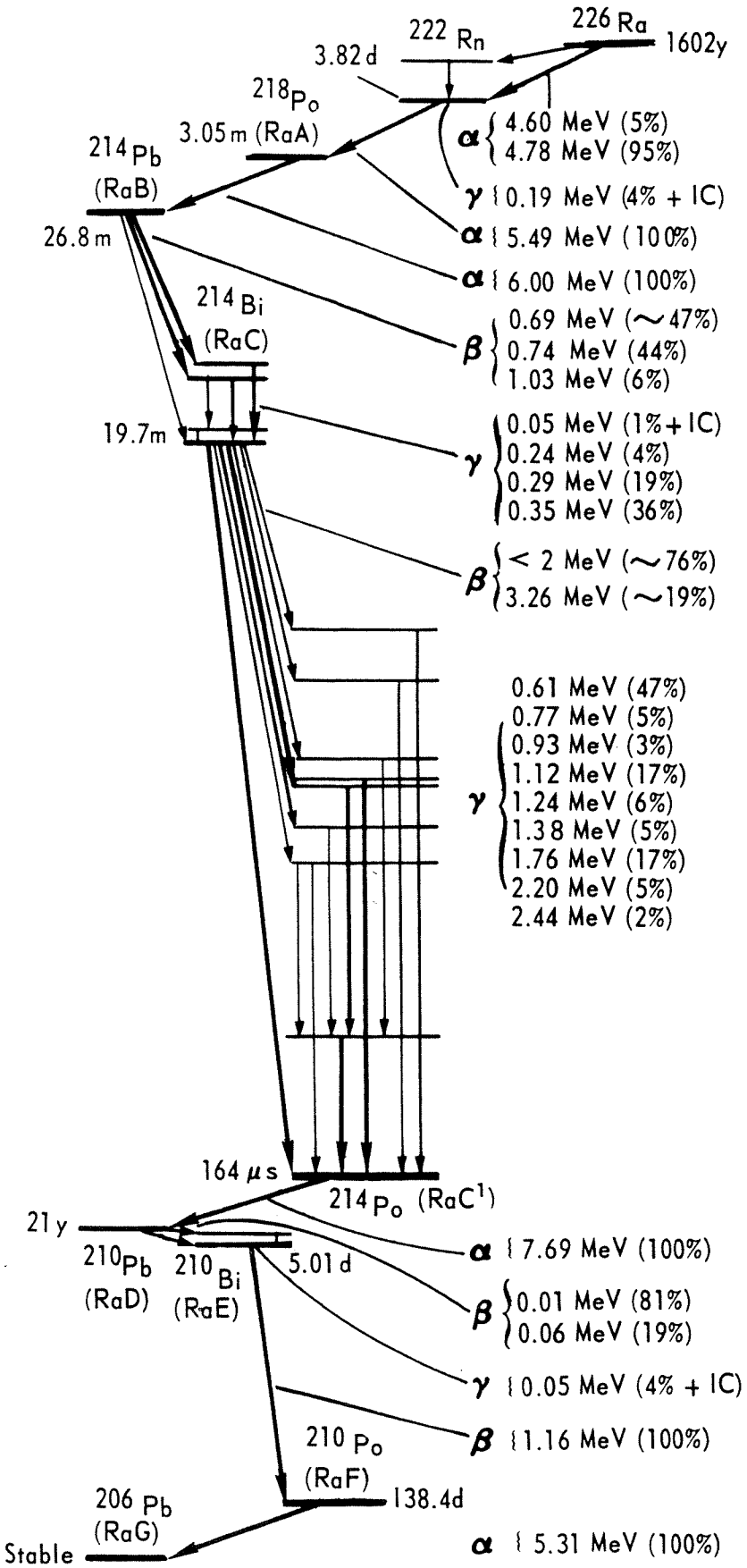
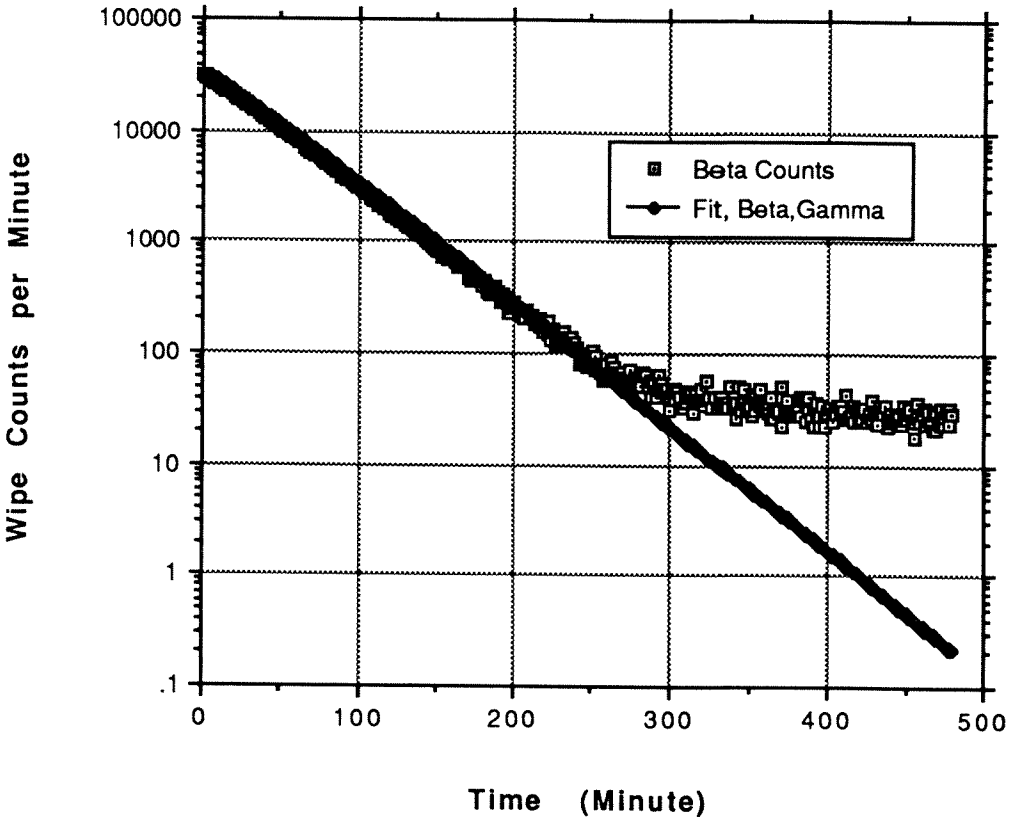

226 Ra
1602y
222 Rn
3.82 d
218 Po
3.05 m (RaA)
214 Pb
(RaB)
26.8 m
214 Bi
(RaC)
19.7m
164 μs
214 Po (RaC1)
21 y
210 Pb
(RaD)
210 Bi
(RaE)
5.01 d
210 Po
(RaF)
138.4d
206 Pb
(RaG)
Stable
α 4.60 MeV (5%)
4.78 MeV (95%)
γ 0.19 MeV (4% + IC)
α 5.49 MeV (100%)
α 6.00 MeV (100%)
β 0.69 MeV (~47%)
0.74 MeV (44%)
1.03 MeV (6%)
γ 0.05 MeV (1% + IC)
0.24 MeV (4%)
0.29 MeV (19%)
0.35 MeV (36%)
β < 2 MeV (~76%)
3.26 MeV (~19%)
γ 0.61 MeV (47%)
0.77 MeV (5%)
0.93 MeV (3%)
1.12 MeV (17%)
1.24 MeV (6%)
1.38 MeV (5%)
1.76 MeV (17%)
2.20 MeV (5%)
2.44 MeV (2%)
α 7.69 MeV (100%)
β 0.01 MeV (81%)
0.06 MeV (19%)
γ 0.05 MeV (4% + IC)
β 1.16 MeV (100%)
α 5.31 MeV (100%)

Figure 3

BETA, GAMMA EMISSION DATA, 226-1.1-6



ALPHA EMISSION DATA, 226-1.1-6

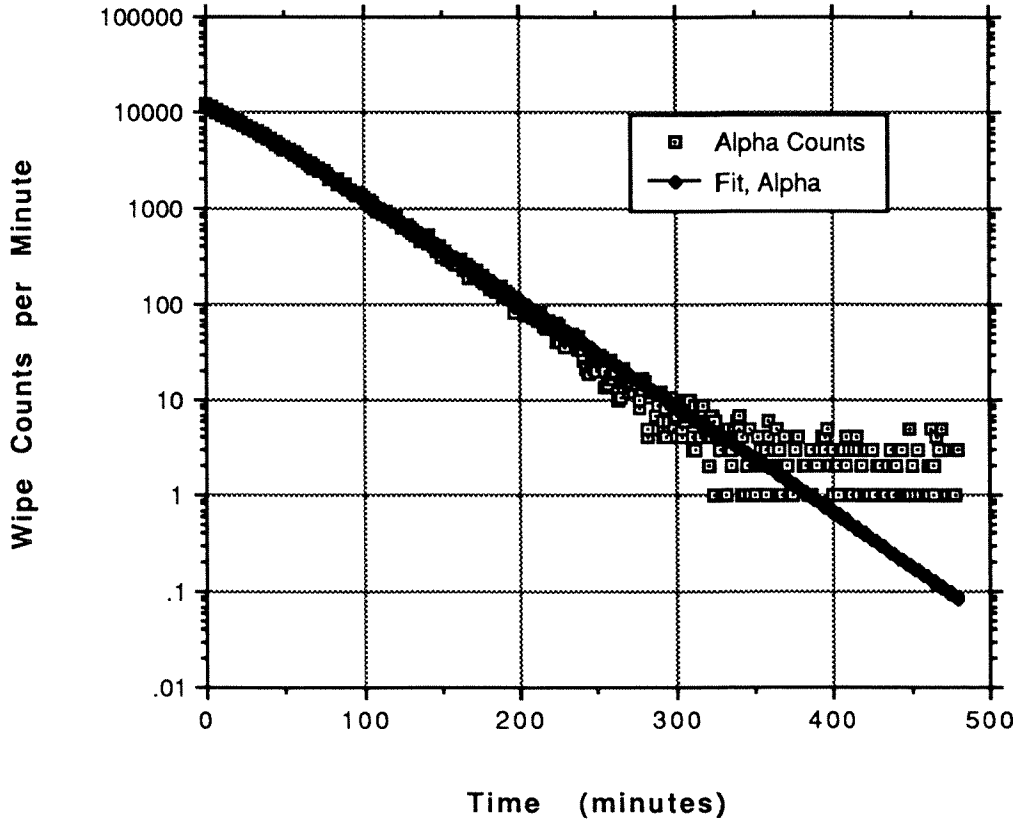


Figure 4

BETA, GAMMA EMISSION DATA, 226-1.1-6